



UNIVERSITI PUTRA MALAYSIA

**MAGNETORESISTANCE OF CuCoNi, CuCo AND AlFeNi GRANULAR
THIN FILMS PREPARED BY RF MAGNETRON SPUTTERING**

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**By
YU ONG SING**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
In Fulfilment of the Requirement for the Degree of Master of Science**

June 2002



Abstract of the thesis presented to the Senate of Universiti Putra Malaysia in
fulfillment of the requirements for the degree of Master of Science

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Chairman: Professor Dr. Abdul Halim Shaari

Faculty: Science and Environmental Studies

In this project, a series of Cu-based and Al-based granular thin films have been prepared using the RF magnetron sputtering system. The effect of the temperature, magnetic field and annealing temperature on the magnetoresistance (MR) of the samples prepared has been discussed. It has been demonstrated that the MR is composition, thickness and temperature dependent.

For Cu-based sample which has been prepared under ambient temperature, the highest MR of 1.35% is observed for sample $\text{Cu}_{80.46}\text{Co}_{19.54}$ (CC6). The samples with different compositions of magnetic entities show the different optimum thickness that would give the highest MR. The samples $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ and $\text{Cu}_{75.63}\text{Co}_{19.56}\text{Ni}_{4.81}$ have the maximum MR when the deposition time is 80 minutes whereas for sample $\text{Cu}_{80.46}\text{Co}_{19.54}$, the maximum occurs when the deposition is 60 minutes.

Upon annealing, the MR of samples also changes. In this study, all of the Cu-based samples prepared at ambient temperature have been annealed at the temperature of 300°C, 350°C, 400°C, 450°C for 10 minutes in the argon environment. Among the samples, $\text{Cu}_{80.46}\text{Co}_{19.54}$ shows the highest MR of 1.8% when it is annealed at 450°C.

From this study, it is also found that for the same thickness (Cu-based samples), there is a general tendency for MR to increase with additional Ni content. Except for the sample annealed at 450°C, the highest MR occurs in the sample with 7.97% of Ni.

The samples $\text{Cu}_{77.18}\text{Co}_{21.82}$ (CC7), $\text{Cu}_{74.67}\text{Co}_{25.33}$ (CC8), $\text{Cu}_{71.31}\text{Co}_{28.69}$ (CC9), $\text{Cu}_{72.21}\text{Co}_{23.01}\text{Ni}_{4.78}$ (CC7N1), $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ (CC6N2), $\text{Cu}_{72.53}\text{Co}_{13.24}\text{Ni}_{14.23}$ (CC4N4) are prepared at 100°C. It has been shown that when the percentage of the matrix remains the same, the composition of the magnetic entities will influence the MR. In this study, $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ has the highest MR of ~2.5%.

From the XRD patterns, it is obvious that all samples consist of Cu(111) and Co(111) texture. Higher MR is observed for the sample when the formation of the Cu(111) texture is dominant.

The MR of AlFeNi granular films is also temperature and thickness dependent. Among the three samples, $(\text{Al}_{71.25}\text{Fe}_{15.34}\text{Ni}_{13.41})$, $\text{Al}_{64.64}\text{Fe}_{21.54}\text{Ni}_{13.82}$,

$\text{Al}_{70.46}\text{Fe}_{21.86}\text{Ni}_{7.68}$), $\text{Al}_{71.25}\text{Fe}_{15.34}\text{Ni}_{13.41}$ and $\text{Al}_{64.64}\text{Fe}_{21.54}\text{Ni}_{13.82}$ have the optimum sputtering time of 230 minutes with the MR of ~0.55% and ~0.35% respectively.

The XRD patterns of the Al-Based samples depict that Fe(110) and Ni(111) textures are dominant among the samples. The intensity of the magnetic entities is higher than that of the Al(111) matrix and for the sample AlFe_2Ni_2 the Al(111) texture has the lowest intensity. When the magnetic entities increases, the XRD peak moves to higher angle. This shows that with the increment of the magnetic entities in the sample, there are more Fe and Ni atoms diffused in the Al matrix leading to the lattice shrinkage.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia bagi memenuhi keperluan Ijazah Master Sains

MAGNETORINTANGAN BAGI SAPUT TIPIS CuCoNi, CuCo DAN AlFeNi YANG DISEDIAKAN DENGAN MENGGUNA TEKNIK PERCIKAN MAGNETRON FREKUENSI RADIO (RF)

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Dalam projek ini, sampel-sampel yang berasaskan Cu dan Al telah disediakan dengan menggunakan teknik percikan magnetron frekuensi radio. Kesan suhu, medan magnet dan sepuhlindapan pada magnetorintangan bagi sampel-sampel yang disediakan telah dikaji. Magnetorintangan bagi sampel-sampel yang disediakan adalah bergantung kepada komposisi, ketebalan dan suhu.

Sampel-sampel yang berasas Cu yang disediakan pada suhu bilik menunjukkan nilai magnetorintangan tertinggi pada sampel $\text{Cu}_{80.46}\text{Co}_{19.54}$ (CC6), iaitu sebanyak 1.35%. Sampel yang mempunyai komposisi entiti magnet yang berlainan akan menunjukkan nilai maksimum magnetorintangan pada ketebalan yang berlainan. Siri $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ dan $\text{Cu}_{75.63}\text{Co}_{19.56}\text{Ni}_{4.81}$ menunjukkan nilai magnetorintangan tertinggi pada sampel yang dipercikan selama 80 minit, manakala

bagi siri $\text{Cu}_{80.46}\text{Co}_{19.54}$ nilai maksimum berlaku pada sampel yang dipercikan selama 60 minit.

Selepas sepuhlindapan, nilai magnetorintangan bagi sampel-sapel juga berubah. Dalam kajian ini, semua sampel, berasas Cu, yang disediakan pada suhu bilik telah disepuhlindap pada suhu 300°C , 350°C , 400°C dan 450°C selama 10 minit. Sepuhlindapan dijalankan dalam persekitaran gas argon (argon environment). Di antara sampel-sampel, $\text{Cu}_{80.46}\text{Co}_{19.54}$ yang disepuhlindah pada suhu 450°C menunjukkan nilai magnetorintangan yang tertinggi, iaitu sebanyak 1.8%.

Dalam kajian ini, juga didapati untuk ketebalan yang sama (bagi sampel yang berasas Cu), nilai magnetorintangan akan bertambah dengan tambahan Ni pada sampel. Kecuali pada sampel yang disepuhlindap pada suhu 450°C , nilai magnetorintangan tertinggi berlaku pada sampel yang mengandungi 7.97% Ni.

Sampel-sampel $\text{Cu}_{77.18}\text{Co}_{21.82}$ (CC7), $\text{Cu}_{74.67}\text{Co}_{25.33}$ (CC8), $\text{Cu}_{71.31}\text{Co}_{28.69}$ (CC9), $\text{Cu}_{72.21}\text{Co}_{23.01}\text{Ni}_{4.78}$ (CC7N1), $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ (CC6N2), $\text{Cu}_{72.53}\text{Co}_{13.24}\text{Ni}_{14.23}$ (CC4N4) telah disediakan pada suhu 100°C . Kajian menunjukkan apabila peratusan matriks dikekalkan, komposisi entiti magnet yang dicampur akan mempengaruhi nilai magnetorintangan. Dalam kajian ini, $\text{Cu}_{72.41}\text{Co}_{19.62}\text{Ni}_{7.97}$ menunjukkan nilai magnetorintangan yang tertinggi, iaitu sebanyak ~2.5%.

XRD menunjukkan semua sampel yang berasas Cu, mempunyai puncak Cu(111) dan Co(111). Kajian mendapati apabila Co(111) dominan pada sampel, nilai magnetorintangan adalah lebih tinggi.

Nilai magnetorintangan bagi saput tipis AlFeNi juga bergantung kepada suhu dan ketebalan. Diantara tiga sampel, ($\text{Al}_{71.25}\text{Fe}_{15.34}\text{Ni}_{13.41}$, $\text{Al}_{64.64}\text{Fe}_{21.54}\text{Ni}_{13.82}$, $\text{Al}_{70.46}\text{Fe}_{21.86}\text{Ni}_{7.68}$), $\text{Al}_{71.25}\text{Fe}_{15.34}\text{Ni}_{13.41}$ and $\text{Al}_{64.64}\text{Fe}_{21.54}\text{Ni}_{13.82}$ mempunyai masa percikan optimum selama 230 minit dengan nilai magnetorintangan $\sim 0.55\%$ dan $\sim 0.35\%$ masing-masing.

XRD bagi sampel-sampel yang berasas Al menunjukkan Fe(110) dan Ni(111) adalah dominan. Corak XRD menunjukkan keamatan entiti magnet adalah lebih tinggi daripada matrix Al(111). Sampel AlFe_2Ni_2 menunjukkan keamatan Al(111) yang terendah. Apabila entiti magnet bertambah, puncak XRD beranjak ke sudut yang lebih tinggi. Ini menunjukkan dengan bertambahnya entiti magnet pada sampel, lebih banyak atom Fe dan Ni akan resap ke dalam matriks dan menyebabkan pengecutan kekisi.

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I certify that an Examination Committee met on 5th July 2002 to conduct the final examination of Yu Ong Sing on his Master of Science thesis entitled "Magnetoresistance Of CuCoNi, CuCo And AlFeNi Granular Thin Films Prepared By RF Magnetron Sputtering" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.



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LIST OF ABBREVIATIONS/SYMBOLS

ASM	Artificially structured material
AMR	Anisotropic magnetoresistance
CIP	Current in plane
CPP	Current perpendicular to plane
CVD	Chemical vapour deposition
EDX	Energy dispersive X-ray analysis
EMA	Effective medium approach
GMR	Giant magnetoresistance
FM	Ferromagnetic
MBE	Molecular beam epitaxy
MR	Magnetoresistance
OMR	Ordinary magnetoresistance
PVD	Physical vapour deposition
XRD	X-ray diffraction
S	Finite cross section
L	Length
G	Conductance
σ	Conductivity
σ_c	Core's conductivity
σ_s	Shell's conductivity
σ_e	Effective conductivity
λ_{mfp}	Mean free path

λ_{sd}	Spin diffusion length
K_F	Fermi wave vector
ρ	Resistivity
$\Delta\rho$	Change of resistivity
K_x	Demagnetizing factor
\vec{J}	Current density
\vec{E}	Electric field
V_f	Volume fraction of the coated grain
V_p	Percolation volume fraction
Y	Sputtering yield
v	Electron velocity
m	Electron mass
\vec{B}	Magnetic field strength

CHAPTER 1

INTRODUCTION

1.1 Magnetic Materials

Magnetism has been known for thousands of years, the word *magnetic* comes from the Greeks and referred to an area in the Mediterranean known as Magnesia where magnetic lodestones were found. Improved smelting techniques, advances in metallurgy, and a greater understanding of the behaviours of magnetic metals over the past century means that today researchers can choose from among a wide variety of magnetic metals and alloy to study. An example: the most familiar magnet, the household refrigerator magnet, is a relatively weak magnet formerly made from an alloy of aluminium, nickel, and cobalt plus other materials, and hence referred to as an Alnico magnet (from the periodical table designations of the magnet, three essential elements besides iron are Al, Ni and Co). Today such magnets comprise of barium, iron and oxygen. It turns out that a careful selection of the percentages of various metals in an alloy can optimise certain characteristics of the alloy.

Magnetic materials represent a multibillion-dollar industry. A fundamental understanding of magnetism in alloys has the potential to influence the design of magnetic materials for applications ranging from power generation to data storage. Magnetism has a profound effect on many alloy properties such as phase stability, thermal expansion, and electrical conductivity.

Magnetism is a consequence of electron spin. In metal the same electrons that give rise to cohesion (the energy that holds the crystal together) can, if they are reasonably well localised about atomic sites, i.e., d-electrons in cobalt, nickel and

iron, also give rise to magnetism. Magnetism occurs when it is energetically favourable on the atomic sites to have an excess of electrons of one spin; this spin imbalance gives rise to magnetic moment associated with individual atoms. In a ferromagnet, the local magnetic moment point in the same direction resulting in a macroscopic magnetic moment. In this situation, there are more electrons with spins that point parallel to the spin axis of quantization than electrons whose spins point antiparallel. In an antiferromagnetic, an equal number of moment point up and down in an ordered arrangement, resulting in no net microscopic magnetic moment.

1.2 Giant Magnetoresistance

Magnetoresistance was discovered by British physicist William Thomson in 1856. However, it could not be explained until the development of quantum mechanics in the 1920s. Basically, a magnet held near certain metals causes their atoms to tilt. The tilted atoms present larger obstacles to passing electrons, thus presenting higher resistance. The giant phenomenon stems from the additional fact that electrons can spin up and down. To change the resistance, it is necessary to construct a material in which one of these electron types can pass through easily, aided by the external magnetic field.

In GMR materials, the layers are used to form a sandwich of magnetic layers, with a non-magnetic material as the filling. In this way, each successive magnetic layer is naturally magnetised in the opposite direction, in the same way as the poles of bar magnets line up in opposite directions when placed against each other. When electric current is passed through such a sandwich, both up and down electrons encounter many obstacles. With the aid of applied magnetic field, all the magnetism

in the layers is forced to line up and down in the same direction as the electrons in the current to avoid the obstacles, creating a short-circuit effect and a large drop in resistance.

Giant magnetoresistance may be most easily understood through a simple optical analogy (Figure 1.1). If a beam of unpolarized light is directed through a pair of polarizers, the total transmitted intensity can be modulated by rotating the polarisers with respect to each other. The first polarizer scatters all but one polarization of light, and the second polarizer either transmits that the first polarization (0°) or block it (90°).

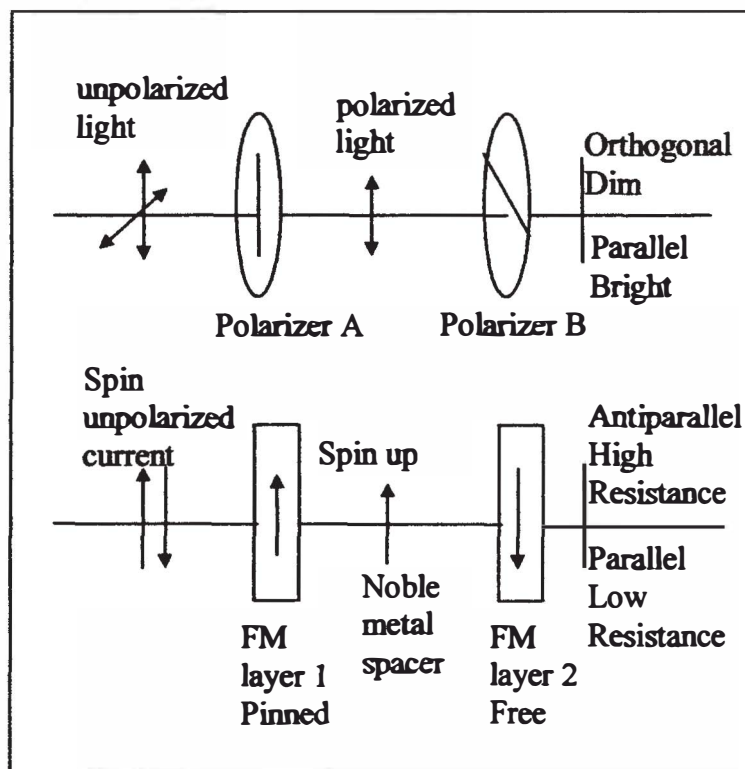


Figure 1.1: Explanation of GMR by Analogy

For GMR, electrons take the place of photons, thin ferromagnetic materials act as electronic polarizers, and the polarization is in terms of spin rather than electric field. In this context, the film is assumed to consist of three thin layers: two ferromagnetic layers separated by a noble metal spacer layer. The polarization axis is provided by the magnetization of the ferromagnet through spin dependent scattering. Conduction electrons with spin parallel to the layer magnetization are scattered weakly close to the layer, carrying current more effectively and leading to low resistance, and those with spin antiparallel to the magnetization are scattered strongly, leading to high resistance. Spin dependent scattering of this sort has been postulated to be the dominant resistance mechanism in bulk ferromagnets since the work of Mott (Mott, 1964).

In fact GMR effect is not exclusive to multilayers and sandwich structure. Xiao has shown that GMR phenomenon also occurs in granular materials in which magnetic single domain particles are embedded in a metallic matrix (Xiao et.al., 1992). The magnetic matrix may consist of Ag, Cu, Au, Cr, etc., whereas the magnetic particles are Co, Fe, and Ni. Granular systems have drawn the worldwide attention due to its unique properties. MR in granular systems is essentially isotropic and the granular films are relatively easier to prepare when compared to multilayers.

1.3 The Objectives of the Project

Firstly, the aim of this project is to determine the suitable parameters and conditions for producing high quality CuCo, CuCoNi and AlFeNi granular thin films using RF magnetron sputtering. Then the composition, thickness and temperature dependence of GMR are studied. The effect of annealing temperature upon GMR